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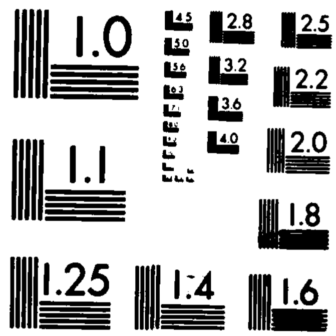
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AD-A143 295

FINAL REPORT ON

ONR CONTRACT

N00014-82-K-0209

"VISIBLE AND NEAR-INFRARED DISSOCIATION LASERS"

Prepared for

Dr. M. B. White
Office of Naval Research
495 Summer St.
Boston, MA 02210

Prepared by

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July 1984

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Experiments have been conducted in which a tunable, injection-locked cadmium monoiodide (CdI) laser has been demonstrated. Over a 50 Å spectral region centered at 657 nm, complete locking of the slave amplifier occurs for injected laser intensities of 5 W - cm^{-2}. Also, an electron beam pumped I_2 green excimer amplifier at 506 nm has been developed. Coupling between the UV and green bands of I_2 has been shown to be strong and the small signal gain coefficient at 506 nm is > 5% - cm^{-1}.

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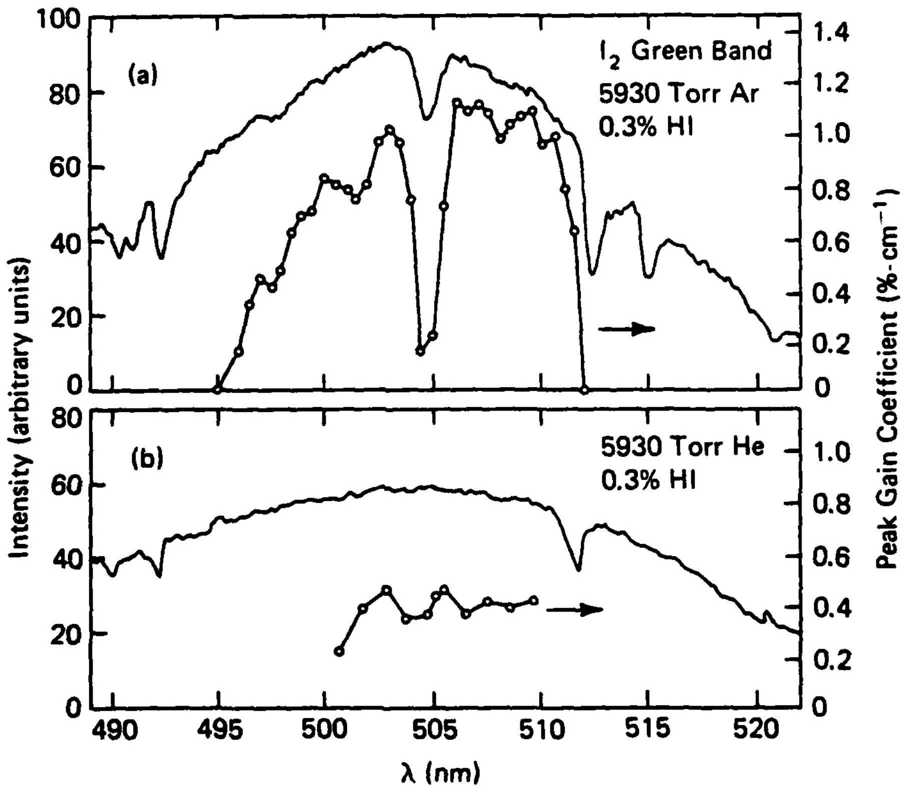
Our research in the study of tunable molecular lasers has been quite productive, specifically with regard to the work on the I_2 green excimer laser and the cadmium-monohalide discharge laser.

A. I_2 Green Amplifier

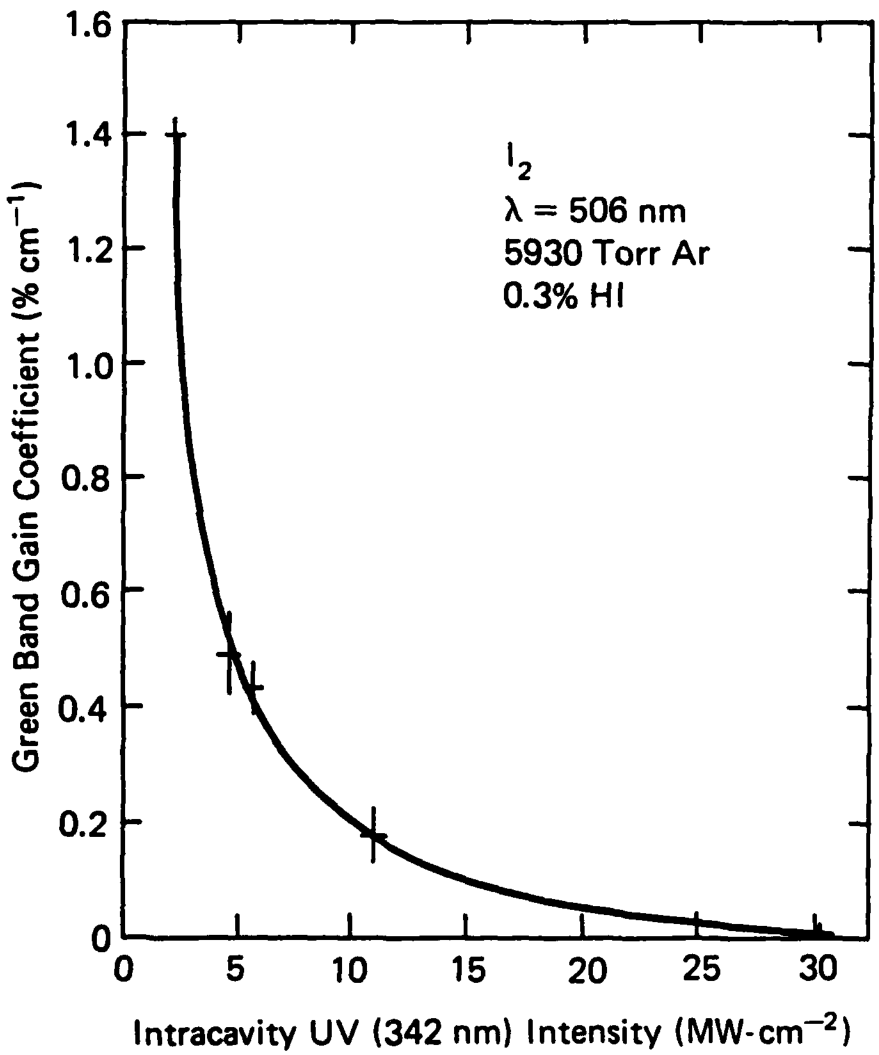
During this time, we have extensively investigated the optical properties of this bound-to-free transition in diatomic iodine under electron beam excitation of dilute mixtures of hydrogen iodide in high pressure rare gas buffers. Using a coaxial diode that was built to mate with a Febetron 706 electron beam generator as an excitation source, we have observed spontaneous emission and amplification on this transition centered at 505 nm with a bandwidth of ~ 150 Å. The details of this work have been reported in our attached paper: Gain on the green (504 nm) excimer band of I_2; Appl. Phys. Lett. 43, 539 (1983). Figure 1 shows the gain and emission profiles for this green band in argon and neon gas diluents.

One interesting aspect of the work on I_2 that was discussed only briefly in this article is the relationship between the upper molecular states of the green band and that for the 342 nm emission (the D' state). It was found that a strong inverse relationship exists between the gain measured on the 505 excimer band and the intensity of stimulated emission on the 342 nm band. Under the nominal conditions used to obtain ~ 1.2% cm^{-1} amplification at 506 nm in I_2, the 342 nm $D' \rightarrow A'$ transition superfluoresces with an intensity of 1.8 MW cm^{-2}. When this is increased to 30 MW cm^{-2} using a high Q cavity to enhance the circulating UV flux, the gain at 506 nm is completely extinguished. This is shown in Figure 2 which is a plot of gain on the I_2 green band at 506 nm versus intracavity flux on the UV laser transition. Note the steep slope of the curve at 1.8 MW - cm^{-2} which supports our earlier calculations that the 506 nm



100
80
60
40
20
0
(a)
I_2 Green Band
5930 Torr Ar
0.3% HI
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
80
60
40
20
0
(b)
5930 Torr He
0.3% HI
1.0
0.8
0.6
0.4
0.2
0
490
495
500
505
510
515
520
λ (nm)
Intensity (arbitrary units)
Peak Gain Coefficient (%-cm^{-1})

Figure 1.



1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
Green Band Gain Coefficient (% cm^{-1})
I_2
λ = 506 nm
5930 Torr Ar
0.3% HI
0
5
10
15
20
25
30
Intracavity UV (342 nm) Intensity (MW-cm^{-2})

Figure 2.

gain coefficient at this intensity is only ~ 20% of its small signal value. Though we do not as yet have the complete picture on the relationship between the upper states of these bands concerning energy defect and coupling, it is likely that suppressing the 342 nm UV superfluorescence will lead to a dramatic increase in the amplification measured in the green.

To obtain a measure of the ultimate gain possible from this visible excimer, more information is needed about this coupling as well as experiments designed to measure the extraction efficiency on this band. A high powered (> 10 MW - cm^2) probe signal from an excimer pumped dye laser could be used to study the effect of visible amplification and UV (342 nm) emission versus probe signal intensity. These experiments could best be carried out using longer pulse e-beam pump devices which transfer greater energies to the excited manifold of states in I_2 than can be realized with our present source. This data should help provide a more complete idea of the viability of this system as a tunable green amplifier.

B. CdI Discharge-Pumped Laser

Tunable, efficient sources of coherent radiation in the visible and near infrared are attractive for a variety of applications, including photochemistry communications and countermeasures. This requires gain media with a large degree of homogeneous broadening such that the energy stored in the upper laser level can be efficiently extracted at any wavelength in the gain spectrum.

As a demonstration of the tunability of new laser media in the visible, an injection locking experiment has been performed on a cadmium monoiodide UV-preionized, transverse discharge laser. This laser was recently developed here at the Gaseous Electronics Laboratory and to our knowledge is the only one of its kind in existence.

The injection locking experimental apparatus is shown in Fig. 3. The ~ 1 μs long pulse from a Chromatix CMX-4 flashlamp pumped, tunable dye laser was injected into the CdI discharge cavity through the 1.35% transmitting mirror. The dye laser linewidth was narrowed to 0.3 cm^{-1} with an internal etalon. Part of the dye laser pulse was split off to monitor its intensity with a calibrated vacuum photodiode. All intensities mentioned are those which pass through the input mirror, i.e., the actual intensity injected into the cavity. No attempt was made to mode match in this experiment. The timing circuit triggered the fast CdI discharge (120 ns FWHM current pulse) sometime during the longer dye laser pulse. The output spectrum through the 0.27% transmitting mirror was recorded with a PARC model 1450 optical multichannel analyzer using a model 1454 intensifying head coupled to a 0.6 m Hilger-Engis spectrometer. The spectrometer was operated in first order and the slits were adjusted to provide resolution of 1.4 Å.

Figure 4 shows the integrated output intensity dependence of the CdI laser on the injected wavelength for a constant injected intensity of ~ 5 W cm^{-2}, which was the maximum intensity used in this experiment. The upper curve is the B → X fluorescence spectrum of CdI between 650 and 662 nm and the lower curve is a composite of the output spectra for nine different injected wavelengths. Each line is also the sum of ten shots to average out shot-to-shot variations. Between 655 and 660 nm, locking is complete at 5 W cm^{-2} and tuning over this did not show any region where locking could not be achieved. The integrated output intensity varied by only a factor of 3 over this range. Outside this region, locking is always incomplete at 5 W cm^{-2} although various amounts of energy could be extracted at the injected wavelength.

The effect of the injected dye laser intensity on the CdI laser output spectrum is seen in Fig. 5 where the dye laser wavelength is set at 653.1 nm.

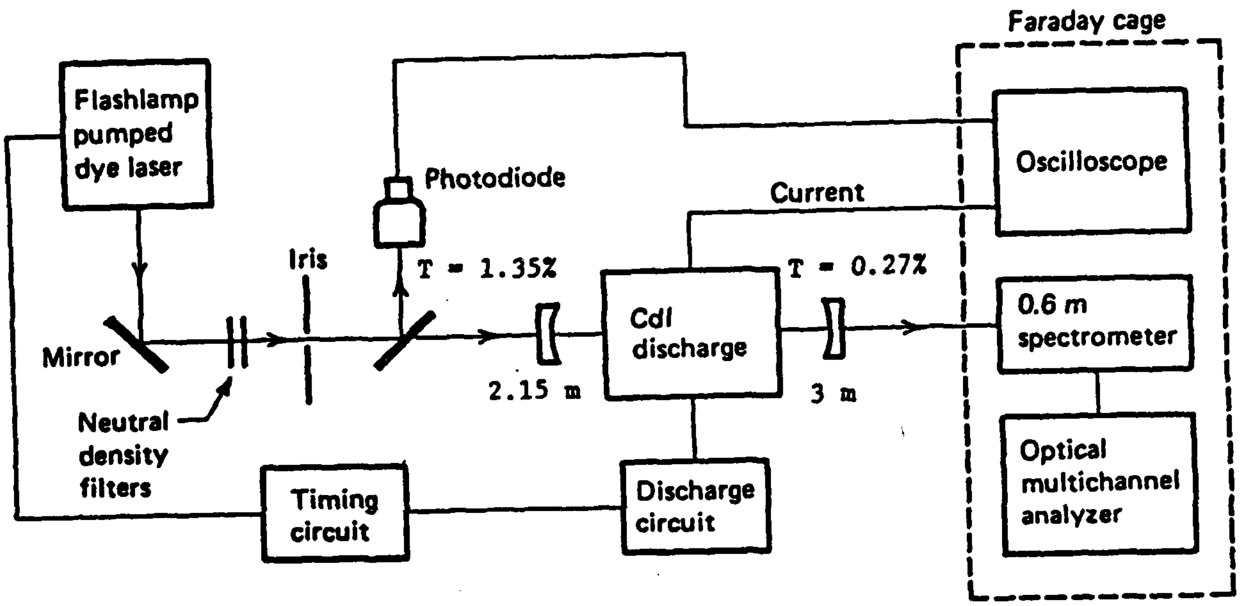
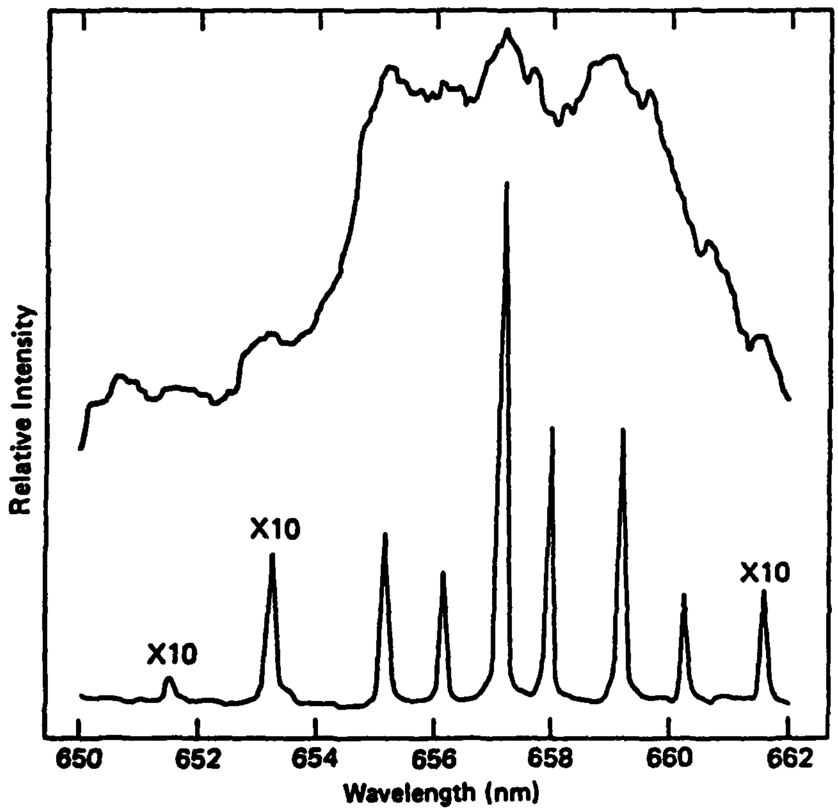
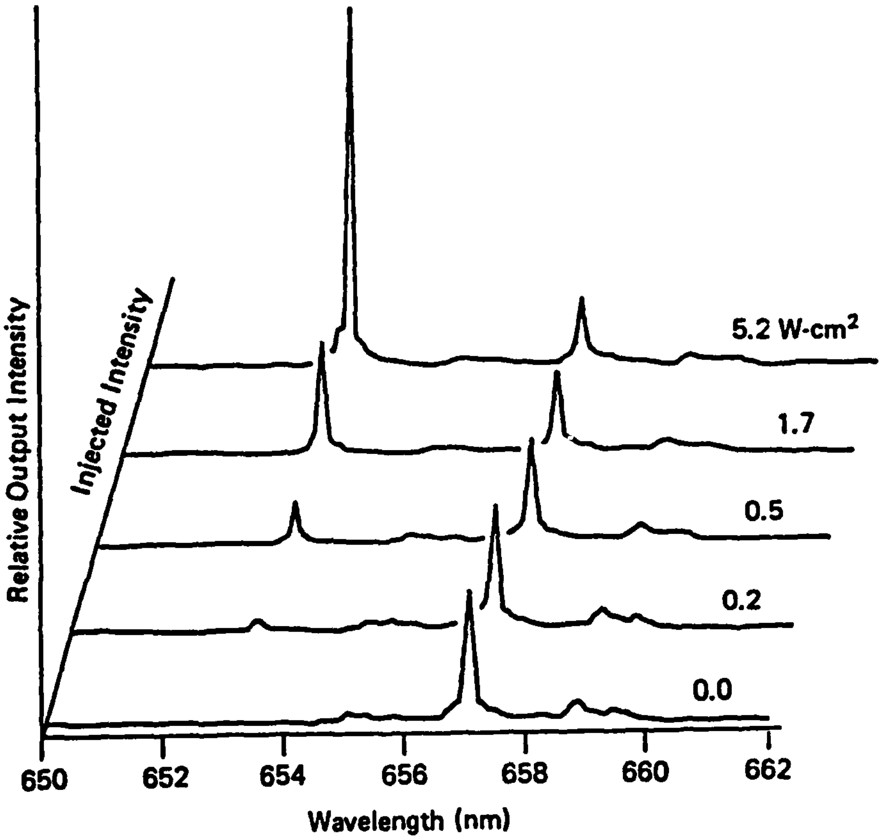

Faraday cage
Flashlamp pumped dye laser
Photodiode
Oscilloscope
Current
Iris
T = 1.35%
T = 0.27%
Mirror
CdI discharge
0.6 m spectrometer
2.15 m
3 m
Neutral density filters
Optical multichannel analyzer
Timing circuit
Discharge circuit

Figure 3.



Relative Intensity
X10
X10
X10
650
652
654
656
658
660
662
Wavelength (nm)

Figure 4.

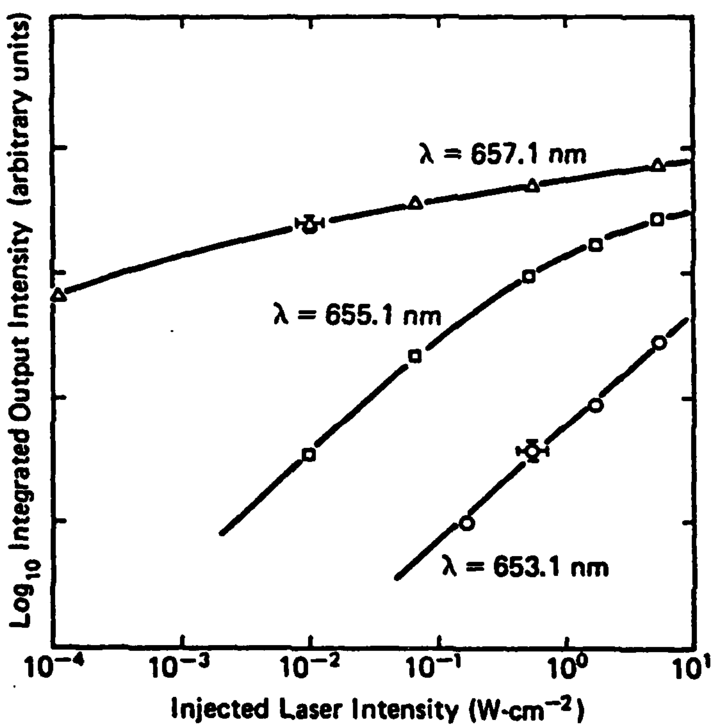


Relative Output Intensity
Injected Intensity
5.2 W-cm²
1.7
0.5
0.2
0.0
650
652
654
656
658
660
662
Wavelength (nm)

Figure 5.

As the injected intensity increases, more energy is extracted at 653.1 nm and less on the naturally lasing line at 657.1 nm. For this case, locking is not quite complete at the maximum intensity of 5 W cm^{-2} although the majority of the energy is emitted at 653.1 nm. This data is plotted graphically in Fig. 6, along with data at two other wavelengths: 655.1 nm and 657.1 nm. As shown in this graph, when injecting at 653.1 nm, energy begins to be extracted at the injected wavelength for intensities of only $\sim 10^{-2}$ W - cm^{-2} and the output increases exponentially up to 5 W cm^{-2}. When injecting at 655.1 nm, energy begins to be extracted at intensities of 10^{-3} W cm^{-2} and the output increases until complete locking occurs at $\lesssim 1$ W cm^{-2}. Beyond that, a less than linear increase is seen. The increase is always less than linear at 657.1 nm but there is more than an order of magnitude improvement in the output over the uninjected laser by locking with just 5 W cm^{-2}. The dye laser flux seeding the cavity allows threshold to be reached sooner and therefore more of the stored energy can be extracted.

Two important points should be noted. The first is that only weak intensities ($\lesssim 5$ W cm^{-2}) are needed to achieve complete locking over a 5 nm range between 655 and 660 nm. This is the result of the modest but significant peak gain coefficient of CdI, 2% cm^{-1} at 657.1 nm, which drastically reduces competition from amplified spontaneous emission. The second point is that, as seen in Fig. 6, as the injected intensity increases, the difference in the output at different wavelengths decreases, indicating a high degree of homogeneous broadening. Figure 6 also indicates that with slightly higher injected intensities, complete locking should be achievable across the entire wavelength region of Fig. 4 between 650 and 662 nm. These results are now being prepared for publication.


λ = 657.1 nm
λ = 655.1 nm
λ = 653.1 nm
Log10 Integrated Output Intensity (arbitrary units)
10−4
10−3
10−2
10−1
10⁰
10¹
Injected Laser Intensity (W-cm−2)

PUBLICATIONS UNDER ONR SUPPORT

1. D. P. Greene and J. G. Eden, "Discharge pumped ZnI (599-606 nm) and CdI (653-662 nm) amplifiers," Appl. Phys. Lett., vol. 42, p. 20, January 1983.
2. D. P. Greene and J. G. Eden, "Lasing on the B-X band of cadmium monoiodide (CdI) and ^{114}CdI in a UV-preionized, transverse discharge," Appl. Phys. Lett., vol. 43, p. 418, September 1983.
3. D. P. Greene and J. G. Eden, "Transient absorption spectroscopy of a CdI discharge," (in preparation).
4. D. P. Greene and J. G. Eden, "On the origin of the 454 nm bound-free absorption band in a CdI discharge," (in preparation).
5. D. P. Greene and J. G. Eden, "Injection locked, tunable output from a CdI transverse discharge laser," (in preparation).
6. K. P. Killeen and J. G. Eden, "Gain on the green (504 nm) excimer band of I_2," Appl. Phys. Lett., vol. 43, pp. 539-541, September 1983.
7. K. P. Killeen and J. G. Eden, "Coupling between the green and UV bands of I_2," (in preparation).

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D. P. Greene, Ph.D. Degree in Electrical Engineering, August 1984.

PERSONNEL

J. G. Eden, D. P. Greene, K. P. Killeen

Gain on the green (504 nm) excimer band of I_2

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(Received 15 June 1983; accepted for publication 7 July 1983)

Gain on the green excimer band ($\lambda \sim 504$ nm) of the iodine dimer has been observed in electron-beam-pumped mixtures of Ar (or Ne), and hydrogen iodide. A peak gain coefficient of $\gamma > 1.1\%\ \mathrm{cm}^{-1}$ is measured at 506 nm with a tunable dye laser and the full width at half-maximum (FWHM) of the gain spectrum in Ar diluent is 13 nm. Temporally resolved gain and fluorescence measurements show that the green emission originates from an I_2 ion pair state other than D'. However, strong collisional coupling between the upper states of the UV ($\lambda = 342$ nm) and green bands and superfluorescence on the UV band ($I_{342} \sim 2\ \mathrm{MW\ cm^{-2}}$) appear to limit the green gain coefficient to $< 20\%$ of its small-signal value. Consequently, suppression of superfluorescence on the UV $D' \rightarrow A'$ bands of the homonuclear halogens should lead to a new family of excimer lasers with wavelengths extending from the green into the ultraviolet.

PACS numbers: 42.55.Hq, 33.20.Kf

The well-known molecular iodine (I_2) ultraviolet laser at 342 nm arises from the D' (1432, $^3\pi_{2g}$)$\rightarrow A'$(2431,$^3\pi_{2u}$) transition of the molecule.[1-3] First demonstrated in 1975, this laser has now been pumped by both electron beam[4-7] and optical excitation.[8-11] The collisional processes responsible for I_2 excited state production in the former case have been extensively studied[12] and the predominant mechanism for formation of the lowest lying I_2 ion pair states (I^+I^-) is collisional deactivation of the I^* (^4P) metastables by iodine-containing molecules such as HI, CF_3I, or I_2. Optical pumping of the laser is possible because the I_2 absorption band centered near 190 nm results in the production of excited $D\,^1\Sigma_u^+$ molecules.[13] In the presence of a buffer gas, these molecules collisionally relax to the lowest lying ion-pair state, $D'\,^3\pi_{2g}$, which is the upper energy level for the UV laser. Both the spontaneous emission and laser spectra for the $D' \rightarrow A'$ band exhibit well-developed vibrational structure since the transition is bound$\rightarrow$bound[2]. Output energies up to 13 J have been reported for an I_2 laser optically pumped by the radiation from an open discharge.[10]

In recent work in this laboratory on the iodine-monofluoride laser, a strong green emission band peaked near 504 nm was observed when mixtures of Ar and hydrogen iodide (HI) were irradiated by a relativistic electron beam. Substituting CF_3I for the HI, the green continuum is again present (though considerably weaker) and underlies the long-wavelength portion of the IF 490-nm band. Hemmati and Collins,[14] Guy *et al.*,[3] Baboshin *et al.*,[13] and apparently Shaw *et al.* [see Fig. 1(b) of Ref. 11] have previously observed this I_2 band for rf discharge or optical (ArF laser or atomic iodine emission) excitation, but this is, to our knowledge, the first observation of the emission for electron beam pumping. References 3, 13, and 14 attribute this green continuum to the $D'\,^3\pi_{2g} \rightarrow 2332,\ ^3\Delta_{2u}$ transition of I_2 which is bound$\rightarrow$free (cf. Fig. 1). However, preliminary time-resolved fluorescence measurements indicate that the green and UV bands of I_2 *do not* share the same ion pair upper state (D') but both levels lie close to one another in energy. The experimental evidence which led to this conclusion will be described in detail elsewhere. The transition responsible for the green I_2 band is, nonetheless, bound$\rightarrow$free and so a laser on this transition would be continuously tunable and could correctly be called an excimer laser.

This letter describes the first measurement of net gain on the green excimer band of I_2 in *e*-beam excited mixtures of Ar (or Ne) and HI. The peak gain coefficient (γ) of $1.1\%\ \mathrm{cm}^{-1}$ is observed at 506 nm and the full width of the gain spectrum is ~ 13 nm.

A schematic diagram of the experimental apparatus is shown in Fig. 2. Excitation of the HI/rare gas mixtures is provided by a Febetron 706 *e*-beam generator (10-J stored energy) and a coaxial diode of 23-cm active length. The Febetron delivers 3-ns, 600-kV pulses to a cathode consisting of strips of 25-μm-thick titanium foil situated longitudinally along the inner wall of a 5.5-cm o.d. aluminum tube. Aluminum anodes were fabricated from 7.6-mm i.d. (1-mm wall thickness) tubing by machining the wall (over the central 23-cm region of the tube) down to 250 μm. The desired wall thickness ($\sim 125\,\mu$m) was then obtained by etching the tubes

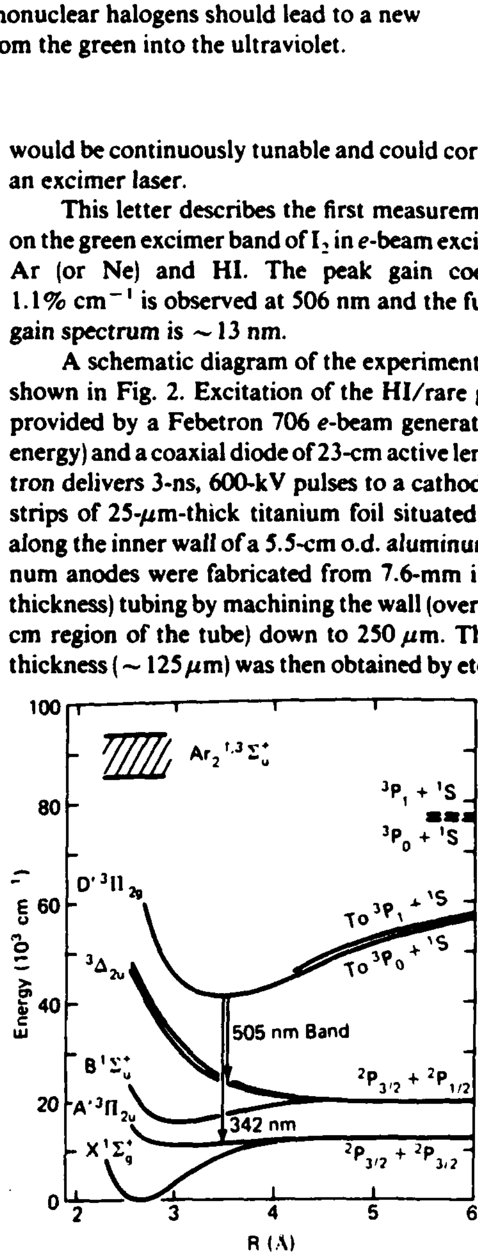

FIG. 1. Partial energy-level diagram for molecular iodine showing the well known $D' \rightarrow A'$ UV band and the green excimer transition (after Refs. 1 and 14). The position of the argon molecular excited states is also indicated. The upper level for the green band appears to be an ion pair state lying above but close to D'.

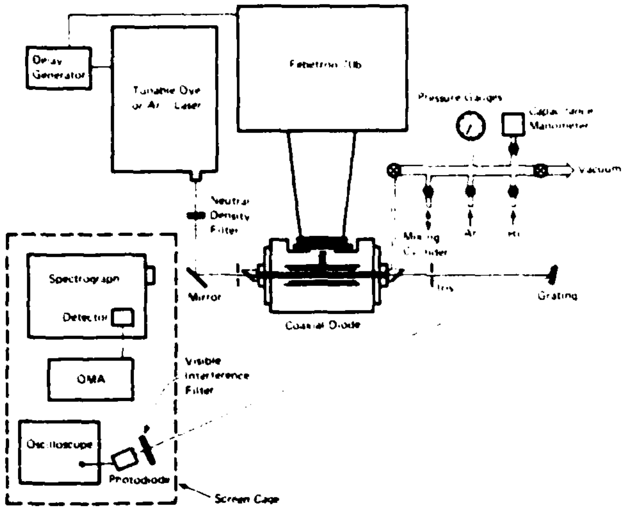

FIG. 2. Schematic diagram of the experimental apparatus. Either an Ar^+ or tunable dye laser provided the probe beam and the grating served to suppress the background spontaneous emission. The turning mirror was removed to obtain the emission spectra.

in a column of 5 M NaOH solution for ~8 min. The active volume of this device is 12 cm^3. A ribbed Lexan insulator sleeve was installed on the cathode stem to prevent flashover to ground. Aluminum Brewster's angle window mounts also served as the means to introduce the gases to the interior of the anode and the diode pressure was maintained at $\sim 10^{-6}$ Torr with an oil diffusion pump system.

Research grade rare gases and electronic grade HI were used as supplied by the manufacturers and the gases were premixed in a steel cylinder. Partial pressures below 100 Torr were measured with a capacitance manometer. Passivation of the system was accomplished by allowing 100 Torr of HI to stand in the vacuum manifold, mixing cylinder, and anode for 24 h.

Fluorescence spectra were recorded by viewing the *e*-beam cell at one end with a Hilger–Engis 0.6-m spectrograph and a PAR optical multichannel analyzer (OMA). The first order dispersion of the spectrograph is ~1.4 nm/mm, giving an overall detection system resolution of ~0.2 nm.

To probe the excited medium for gain, the beam from an Ar ion laser or a flashlamp-pumped, tunable dye laser (Chromatix CMX-4; $\Delta\lambda \sim 0.08$ nm) was directed along the axis of the anode. The steps taken to minimize interference from background fluorescence included placing irises and a diffraction grating in the optical path and locating the vacuum photodiode (S-20 surface) 5 m from the end of the *e*-beam diode. The transmitted probe laser intensity was subsequently displayed on a storage oscilloscope. Low probe intensities (< 10 kW cm^{-2}) were obtained with neutral density filters, thus ensuring that the probe flux did not approach the estimated saturation intensity of the green band. Also, the temporal history of the I_2 UV and green fluorescence was recorded with the same photodiode and interference filters (UV: $T_{max} = 55\%$ at $\lambda_0 = 350$ nm, $\Delta\lambda = 55$ nm; green: $T_{max} = 77\%$ at $\lambda_0 = 500$ nm, $\Delta\lambda = 38$ nm).

Spontaneous emission and gain waveforms from a 0.2% concentration of HI in 5930 Torr of Ar mixture are illustrated in Fig. 3. Actually, the 342-nm transition is superfluorescing, as evidenced by the spectral and temporal narrowing of the emission. (Note that the intensity scales for the green and UV bands shown at the top of the figure are fixed relative to one another.) For a probe laser wavelength of 506 nm, the oscillogram shown in Fig. 3(b) was observed. The strong absorption that is evident throughout the pump pulse is characteristic of electron-beam-excited rare gases[15] and this time period is immediately followed by a gain pulse of 8–10 ns full width at half-maximum (FWHM). This feature is due entirely to gain in the active medium—background spontaneous emission was not observable on this intensity scale.

Probing the excited gases with various Ar^+ and dye laser wavelengths between 495 and 512 nm has resulted in the I_2 green band gain spectra presented in Fig. 4. Each of the gain coefficient data points in the figure is the average of at least three experimental trials. Part (a) shows that, for Ar/HI mixtures, maximum gain is observed at 506 nm ($\gamma = 1.1\%$ cm^{-1}) and the FWHM of the entire profile is ~13 nm. Most of the absorption features that are so prominent in this spectrum [and (b) as well] are too broad to be attributed to excited atomic states and, as noted by Zamir *et al.*,[15] appear to be due to a transition of $Ar_2(^1\Sigma)$ molecules to a Rydberg state. None of the absorptive transitions in (a) or (b) appear to involve the I_2 molecule itself.

Figure 4(b) confirms the assignment of the dominant absorption lines in (a) to the Ar_2 excimer. In this case, Ne is the diluent and the dip in the Ar/HI gain spectrum at $\lambda \simeq 504.5$ nm has vanished. Also, if one corrects for the smaller stopping power of neon as compared to argon, the peak γ in (b) is only slightly less than that for the Ar buffer. Only the central portion of the Ne/HI spectrum was examined in order to confirm the expected disappearance of the 504-nm absorption line. The top curves in (a) and (b) are the fluorescence spectra which have been corrected for the response of the spectrograph and OMA. Aside from the obvious absorption features, these bands are structureless continua, as would be expected.

The dependence of the small-signal gain coefficient at 506 nm on the argon pressure and the gas mixture composition is shown in Fig. 5. The best compromise between optimizing $I^*(^4P)$ formation[12] and minimizing quenching of the upper state occurs for $P_{Ar} \simeq 4000$ Torr and an HI concentration of at least 0.5%.

We note from Fig. 3 that the peak 506-nm gain is measured following the *e*-beam pumping pulse and just after the

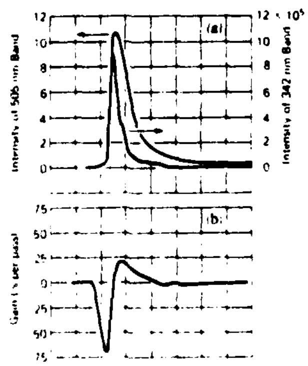

FIG. 3. (a) Fluorescence and (b) gain waveforms for *e*-beam excited I_2. The 342-nm superfluorescing pulse has a full width ~1.1 ns. In (b) the gain observed at $\lambda = 506$ nm for a 0.2% HI in Ar ($p_{TOTAL} \simeq 5930$ Torr) mixture is shown. The peak signal in the oscillogram corresponds to a gain coefficient of 1% cm^{-1}. Gain coefficients as high as 1.2% cm^{-1} have been observed.

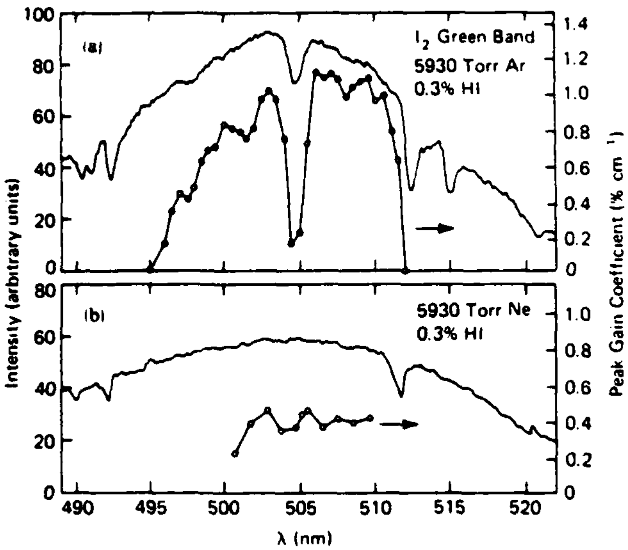

FIG. 4. Spontaneous emission and gain spectra for electron beam excited mixtures of HI and (a) argon or (b) neon. The minimum detectable gain coefficient is $\sim 0.1\%\ cm^{-1}$ and the maximum concentrations of Ar in Ne and Ne in Ar are both less than 1 ppm. The top traces in (a) and (b) are the flourescence spectra as recorded with an OMA.

peak in the 342-nm band intensity. The large optical flux at 342 nm that arises from superfluorescence on the $D'\rightarrow A'$ band strongly depletes the D' population *before* gain is measured on the green transition. Consequently, since the upper states for the UV and 504-nm bands are collisionally coupled, the gain coefficients in Fig. 4 are "saturated." Therefore, under these experimental conditions, the measured gain coefficients are likely well below their *small-signal* values.

To estimate the green (506 nm) small-signal gain coefficient, let the frequency full width and the radiative lifetime τ_r for the $D'\rightarrow A'$ band be $\sim 10^{13}\ s^{-1}$ and 7 ns, respectively.[11,16] Consequently, a conservative estimate[12] for the stimulated emission cross section for this UV band is $\sigma_{SE} \simeq 6\times 10^{-16}\ cm^2$ and the saturation intensity is $I_{sat} = h\nu(\sigma_{SE}\tau_r)^{-1} \simeq 0.14\ MW\ cm^{-2}$. Using a Gen-Tec energy detector, 1 mJ of energy at 342 nm was found to be contained in the 1.1-ns FWHM pulse (so $I_{peak} \sim 1.8\ MW\ cm^{-2}$). Then, if the $D'\rightarrow A'$ transition is homogeneously broadened, the population inversion and γ at peak intensity are both only 8% of their small-signal values. Assuming that the populations of the lower states of the green and UV bands (one dissociative perhaps, $^3\Delta_{2u}$, and the other A', respectively) are negligible, then the inversion on the 504-nm I_2

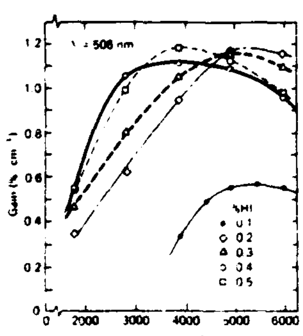

FIG. 5. Dependence of the green gain coefficient on the composition and total pressure of the Ar/HI mixture. The dye laser probe wavelength is 506 nm.

band is also $\sim 1/13$ that present before stimulated emission occurs on the 342-nm transition. Consequently, the presence of a distributed absorber *at 342 nm* in the gain medium may allow the peak gain coefficient *at 506 nm* to rise beyond $5\%\ cm^{-1}$. Viewed from another perspective, saturating the green transition with a dye laser (or HgBr oscillator) pulse should yield reasonable extraction efficiencies.

Although saturated, the gain coefficients measured here are comparable to those for the XeF $(C\rightarrow A)$ band. With the same apparatus, the gain coefficient at $\lambda = 488$ nm was found to be $1.5\%\ cm^{-1}$ for a 1:2:370 mixture of NF_3:Xe:Ar (total gas pressure again $\simeq 6000$ Torr). (In this case, the UV $B\rightarrow X$ band was not superfluorescing.)

In summary, gain on the green excimer band of I_2 has been observed in *e*-beam pumped rare gas/HI mixtures. The room-temperature operation of this system, its large homogeneously broadened gain profile, and attractive spectral position, coupled with the demonstrated energy storage capacity of the I_2 ion-pair excited state manifold, make the band appear promising. Perhaps more importantly, the green band of I_2 is only representative of a family of molecular transitions. Since few excimer gas laser systems exist (and particularly in the visible), one is encouraged to search for similar continua in the other homonuclear halogens Br_2, Cl_2, and F_2.

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